THE EFFECT OF A HIGH ENERGY ELECTRON BEAM ON EXPLOSIVES

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THESIS

THE EFFECT OF A HIGH ENERGY

ELECTRON BEAM ON EXPLOSIVES

by

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ABSTRACT

The effect of a sixty million electron volt beam, produced by a linear accelerator and focused on standard explosives, varying in weight from three to twenty-five milligrams, has been studied. Doses in the range from 1×10^9 rads to 3.2×10^{10} rads completely desensitized lead azide but had no effect on the explosive behavior of lead styphnate. The desensitization tests on explosives consisted of both heat and impact test methods.

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I. INTRODUCTION

With the accelerated pace of technology in nuclear energy, nuclear weapons, and space exploration, and parallel efforts in research and development in the field of ecology, the effects of high energy radiation on standard explosives are extremely important. From the viewpoint of changes in explosive parameters and characteristics, radiation effects on explosive trains could render weapons unpredictable or even unreliable. Furthermore, a desensitization or even dudding effect would greatly aid in finding a solution to the existing problem of recycling obsolete explosives.

This author has previously determined, in another study, that prolonged heat application (approximately three hours at a temperature of 10°C below explosion temperature) would dud lead styphnate. Realizing this method was one of surface effect and chemical change, it was then decided to concentrate on experimentation to determine whether the interaction of high energy electrons (beam energy of 55-65 MeV) with groups of atoms, molecules, and the orbital electrons, would cause changes. It was thought that small chemical changes induced by radiation could lead to large changes in subsequent physical behavior because of modifications of chemical and energy bonds.

Industrial research in the field of radiation effects on primary and secondary explosives and on propellants is being strongly pursued, although much information is classified.

A bibliography produced by the Lawrence Livermore Laboratory [Ref. 4] was most useful. Of particular interest to this author were articles on the subject of initiation of explosives by pulsed electrons, since it was the purpose of this study to try to obtain degradation in explosives; i.e. a dudding effect.

The present study is concerned solely with an investigation into the macroscopic effects caused by electron radiation from a linear accelerator (LINAC). These effects will be presented and discussed. In particular, the relationship between total energy deposited and degradation of the explosive characteristics shall be the major objective of this study.

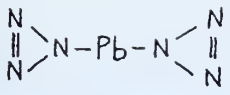
II. THEORETICAL CONSIDERATIONS

Explosives are chemical compounds or mixtures containing energy which, when released, causes the phenomenon known as an explosion. The rate at which the energy is released, rather than the total amount of energy, determines the kind of explosion produced. Characteristics of each type of explosive depend on molecular structures. Thus, ease of detonation by heat or shock, detonation pressure and velocity, brisance (rate of energy release), power and other specifications are a function of the composition and define whether an explosive is an initiator, booster, or a high explosive.

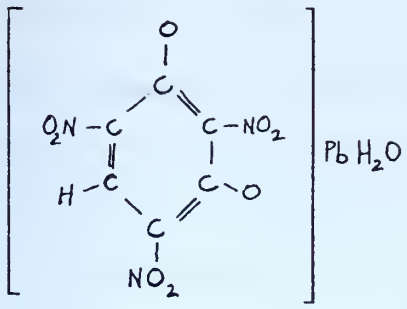
The explosives considered in this study are of varying structure and thus serve different purposes. Thus, lead azide (inorganic explosive compound) and lead styphnate (organic compound) serve as initiators in explosive trains, wherein lead styphnate is normally the first explosive in line and is initiated by heat. Lead azide which is extremely sensitive to shock, such as that created by the initiation of lead styphnate, detonates and initiates the booster, which creates very strong shock waves. They in turn initiate the relatively inert high explosive such as trinitrotoluene (TNT). The booster explosive considered in this study was pentaerythritol tetranitrate (PETN).

Although only one inorganic compound was considered out of a total of five different compounds, the purpose was to

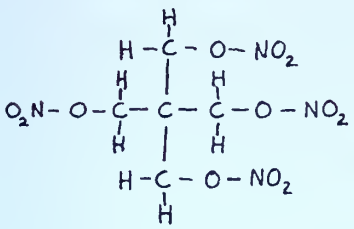
examine types of explosive structures and their behavior after irradiation with high energy electrons. In general, chemical radicals such as the nitro group ($-NO_2$), nitrate group ($-O-NO_2$), and nitramine group ($N-NO_2$), when combined with carbon-hydrogen configurations, form the common explosives. Of the radical groups, the nitrates are more powerful and sensitive (in the sense of shock, temperature, and friction) than the corresponding nitro compounds. The most common of configurations can be classified as being of the benzene ring type or of the carbon chain type. Figure 1 illustrates the structures for the explosives analyzed in this study.



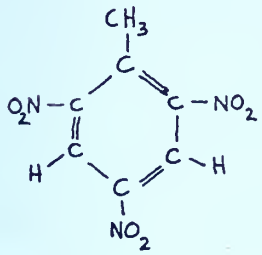
Lead Azide - (1) inorganic compound
(2) non-carbon chain
(3) non-benzene ring



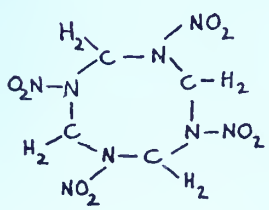
Lead Styphnate - (1) organic compound
(2) benzene ring with nitro groups and ionic bond to PbH_2O



PETN - (1) organic compound
(2) carbon chain with nitrate groups



TNT - (1) organic compound
(2) benzene ring with nitro groups



HMX - (1) organic compound
(2) non-benzene ring
(3) a nitramine ($N-NO_2$ radicals)

Figure 1. Structural formulae of selected explosives.

Some explosive characteristics have been tabulated. Reference [3] does provide some general parameters such as mixtures, explosion temperatures, heat of explosion, and others. In those cases where present literature fails to provide characteristics such as power of explosives, detonation pressure and velocity, and energy released, calculations were made as recommended by J. E. Sinclair [Ref. 11]. A sample of such calculation for bond energy released has been included in Appendix A.

In general, the term "high energy radiation" is taken to include beams of fast electrons. The property of all types of high energy radiation is the large amount of energy carried by each particle (or by a photon) which is much greater than the binding energy of an electron in an atom or the energy holding an atom to other atoms. The large amount of energy per particle allows it to penetrate explosives and possibly disrupt the binding forces between atoms or molecules by the process of ionization, in which electrons are removed from their parent atoms or molecules. This process can cause secondary changes. An electron emitted during ionization may have enough energy to cause secondary ionization and excitation in neighboring atoms. In addition, in the ionization process, the electron which is removed from one molecule will most likely be captured by another molecule. This action results in the production of an ion pair.

Another process is excitation, in which an electron is raised to a higher energy level but remains bound to its

parent nucleus so that the atom or molecule remains neutral. Thus, fast primary electrons produce secondary electrons along their tracks through the sample explosive. These secondary electrons produce, in the neighborhood of the primary electron track, regions of dense ionization. The secondary electrons are really the source of most of the ionization, whereas the primary high energy electrons pass through the sample.

Considering radiation of explosives with high energy electrons, excitation and ionization are by far the most important processes. The removal of an electron, or excitation to a higher energy level, may render molecules unstable and can perhaps cause a chemical reaction resulting in decomposition. Theory indicates that the nature of the resulting processes is extremely complex and unpredictable. The complexity of the explosive bonds, as indicated in Figure 1, supports the fact that the sequence of events from initial ionization to final chemical product occurs in a series of stages which are not completely understood. Even more difficult is the prediction as to products. Irradiation of explosives may result in a variety of chemical changes such as oxidation, nitration, decarboxilation or general degradation of normal reactions and characteristics. For example, the explosives could become less sensitive and a reduction in detonation velocity or pressure could be realized. This study investigated the specific characteristic of complete desensitization.

Of great interest in the theory of explosive composition is the Sponge Effect [Ref. 1]. This effect is pertinent to benzene and acts as a protection against changes caused by radiation. Inspection of Figure 1 reveals that benzene rings are in evidence for the explosives TNT and lead styphnate. Because of the resonant nature of the molecule, the abstraction of an electron still leaves a relatively stable system. Since this stability extends throughout an exposed sample, the benzene molecule acts to absorb much energy. Although Ref. [1] discussed the Sponge Effect for benzene, this author's opinion is that the effect could be extended to compounds containing the benzene ring as their major and central structure. As can be seen in Appendix A, the alternate single and double bonds and the resonance of the benzene ring, figure heavily in the evaluation of bond energy and explosive yield.

The LINAC of the Naval Postgraduate School is a pulsed, three section accelerator with maximum beam energy of 120 MeV. In this experiment only two sections were employed, which gave a beam energy of approximately 60 MeV concentrated in a beam cross-section of about one square centimeter at the target location. The entire explosive sample was contained totally within the beam.

The effect of radiation (bremsstrahlung) in the target explosive was disregarded because of the small sample size and the comparatively large mean free path (normally centimeters) of the resulting photons. Thus, for a beam energy

of 60 MeV, most photons escape from the sample without depositing energy.

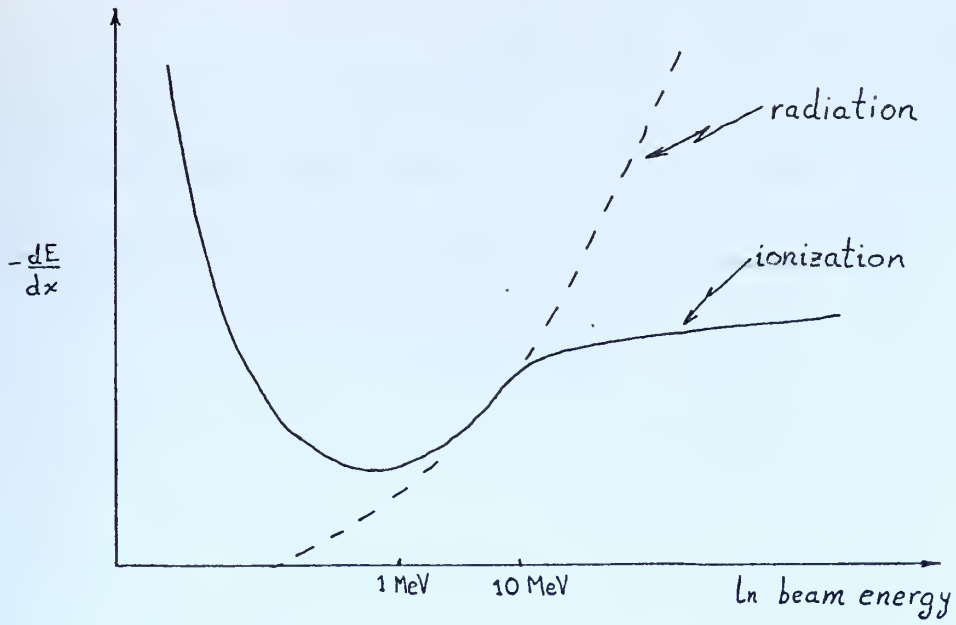


Figure 2. Electron beam energy versus energy loss of incident beam.

Figure 2 indicates the relationship between energy and radiation and ionization energy losses and further illustrates that the ionization loss is reasonably independent of energy above about 10 MeV.

The fact that the flux of the electron beam is not uniform over the 1 cm^2 had to be accounted for. The distribution is approximately Gaussian; therefore, it was desirable to keep the sample confined in such a manner that centering at the maximum flux was possible. In calculations to follow, this author approximated the peak flux (ϕ peak) to be eight times the average flux across the beam profile, based on the measurements of Lesemann [Ref. 5].

The method of obtaining the total energy deposited in each explosive sample was calculated as follows.

The primary electrons passed through the sample and were collected by a Faraday cup. The total charge collected was measured by a vibrating reed electrometer which indicated the voltage across a capacitor charged by the collected electrons.

Since

$$C = \frac{Q}{V} = \frac{ne}{V} ,$$

where

C = capacitance

Q = ne = charge collected by the Faraday cup

e = electron charge

n = number of electrons

V = voltage across C,

then

the number of electrons is

$$n = \frac{CV}{e} .$$

For example, for a setting of 10 volts and 10μf

$$n = \frac{(10 \times 10^{-6}) \times 10}{1.6 \times 10^{-19}} = 6.25 \times 10^{14} ,$$

which is approximately equal to the average fluence, ϕ_{av} , since the beam area is about 1 cm^2.

Therefore,

$$\phi_{peak} = 8 \times \phi_{av} = 8 \times 6.25 \times 10^{14} \frac{e^-}{cm^2}$$

$$\phi_{peak} = 5 \times 10^{15} \frac{e^-}{cm^2} \text{ at the sample.}$$

The dose is then calculated from

$$D = \frac{1}{\rho}\frac{dE}{dx} \times \phi_{peak} ,$$

where

ρ = density of the sample, and

$\frac{dE}{dx}$ = ionization energy loss by the incident electrons $(\frac{MeV}{cm})$.

The assumption that $\frac{1}{\rho}\frac{dE}{dx} \approx$ constant $\approx 2 \frac{MeV\text{-}cm^2}{gm}$ was made; recalling that $\phi_{peak} = 5 \times 10^{15} \frac{e^-}{cm^2}$ for the specific setting of 10 volts and 10μf,

$$D = 2 \frac{MeV\text{-}cm^2}{gm} \times 5 \times 10^{15} \frac{e^-}{cm^2} = 10^{16} \frac{MeV}{gm} .$$

Since

1 MeV = 1.6×10^{-6} ergs, and

1 rad = $10^2 \frac{ergs}{gm}$,

then

$$D = 10^{16} \frac{MeV}{gm} \times \frac{1.6 \times 10^{-6} ergs}{MeV} \times \frac{rad}{10^2 \frac{ergs}{gm}}$$

$D = 1.6 \times 10^8$ rads for 10 volts and 10μf .

This dose was the energy deposited in the explosive sample per single integration with settings of 10 volts and 10μ farads. In order to calculate the total accumulated dose; i.e. the total energy deposited, all integrations were simply added, thus obtaining ϕ_{total} and

$$D_{total} = \frac{1}{\rho}\frac{dE}{dx} \times \phi_{total}$$

The question under consideration, and of primary importance, then related to the amount of energy required to cause

complete desensitization. The first estimation concentrated on the specific explosive bonds (such as only the nitro groups on a benzene ring) and rationalized that at least as much energy should be deposited as would be released by the specific nitro bonds (refer to Figure 1). Considering TNT, for example, there exist three nitro groups. Rupture of these three specific bonds would result in the release of $49.98 \frac{\text{kilo-cal.}}{\text{g-Mole}}$ [Ref. 11]

or $49.98 \times 10^3 \text{cal} \times \frac{1}{227} = 220.0$ cal. (per gram of TNT) released by the $C\text{-}NO_2$ bonds. Since

$$1 \text{ cal} = 4.186 \text{ joule, and}$$

$$1 \text{ joule} = 10^7 \text{ ergs}$$

then

$$220. \frac{\text{cal}}{\text{gm}} \times \frac{4.186 \text{ joule}}{\text{cal}} \times \frac{10^7 \text{ergs}}{\text{joule}} = 9.21 \times 10^9 \frac{\text{ergs}}{\text{gm}}$$

as the $C\text{-}NO_2$ bond energy.

The equivalent energy deposit is thus 9.21×10^7 rads.

Similar calculations for all explosives would lead to estimates of minimum energy to be deposited. Whether this minimum energy would in fact be deposited in the bonds under consideration is uncertain. In most cases, the energy could be deposited throughout the molecule and in a way which is in part random and structure dependent. To deposit energy into an explosive sample which would affect the explosive bonds, it was decided to deposit energy at least ten times or even one hundred times that of the minimum. Hence, the actual estimates of 10^8 rads, 10^9 rads, or even as high as 10^{10} rads seemed reasonable.

III. EXPERIMENTAL PROCEDURES AND EQUIPMENT

The standard explosives used, TNT, HMX, PETN, Pb Styphnate, and Pb Azide were all in powder or crystalline form. Sample sizes varied from 3.3 mg to 25 mg depending on effects expected, the type of test to be performed in the laboratory, and the type of explosive. In addition, sample sizes were dependent on explosive density and thus determined the height of the cylindrical column in the glass tube, which was critical in relation to the 1 cm^2 cross-section of the electron beam. Equipment safety was a primary consideration. An estimate of the energy released and power of detonation was required in order to determine the damage that would be done should a sample explode during a LINAC test and rupture the delicate windows on either the vacuum chamber or Faraday cup. The energy released and detonation pressure and velocity were calculated as indicated in Appendix A. Temperatures for explosion ranged from 211°C for lead styphnate to 475°C for TNT and it was determined that the electron beam would not cause a rise in temperature to such a value. In fact, it seemed that the beam raised the temperature by only a few degrees, as judged by feeling the samples immediately after exposure.

Sample sizes and containers were varied during the study, as were some test methods. However, each variation was made in order to ascertain in a more positive way, whether a sample was dudded by the exposure. The explosive containers

used, as well as a glass dipper, are shown in Figure 3. The outer diameter and inner diameter of the glass tubes were 4.5 mm and 2.5 mm respectively. The progression of usage was from the pocketed dish (in which the sample was emplaced and then a slide epoxied on top), to the longer open end tube, and finally the relatively short open end tube. The initial dish configuration proved to be undesirable in that a shaped charge was formed and also that any gaseous products would be trapped. The long glass tube, although providing the solution to these problems, was later discarded as the sample holder since during the loading process an extremely small residue could cling to the glass and would not be radiated. The small glass container solved that problem since the entire container was less than one centimeter in height and thus totally contained in the beam.

Sample weights were measured to 0.01 mg accuracy with a Metler H-16 balance. The glass tube containing the column of explosive was then placed in a wooden container and carried to the accelerator. Standard explosive safety precautions were always observed.

For all tests, the accelerator target area was arranged so that the beam would pass through the vacuum chamber into the atmosphere, through the target and bakelite card which were placed on an adjustable table, and into the Faraday cup. Figures 4 and 5 illustrate this end station arrangement and also a close-up of the target card without a sample. A Zinc Sulfide screen on the target card aided in the focusing of

the beam. Furthermore, the target card was placed on a table which was movable from the control room in both a horizontal and vertical direction. The monitoring of the target card, beam position and size, and in general the entire end station, was made possible by the use of a T.V. camera in the end station.

Upon completion of accelerator tuning, the spot on the target card was carefully noted, the beam was turned off, and the glass tube containing the sample was carefully scotch-taped to the spot on which the beam had been focused. A Zinc Sulfide paint was then applied over the column of explosive (or over the short glass tube in later tests). This procedure aided in placing the beam directly on the explosive after the target card (with sample) was returned to the stand in the end station. When the end station was secured, then the beam was fine adjusted onto the visible target (see Figure 6). The settings of voltage and capacitance and the final adjustment of beam spotting permitted the gathering of data leading to the dose calculation as previously explained.

The data gathered were put into the following form:

Run #	Accumulated Time (seconds)	Accumulated Dose (e^-/cm^2)	Remarks
1	184	15×10^{15}	= ϕ_{peak} each
2	370	30×10^{15}	integration
'	'	'	
'	'	'	
'	'	'	
		$3.75 \times 10^{17} \frac{e^-}{cm^2}$	= ϕ_{peak} total

The total dose calculations were performed as described in Section II. This method of data gathering and dose calculation was employed for all samples radiated.

Figure 7 illustrates the appearance of the target sample after exposure. Each sample was brought to the explosives laboratory for analysis and for the final test of explosion or no explosion. One sample of lead azide and one sample of lead styphnate were analyzed for unpaired electrons by conducting an electron spin resonance analysis prior to the final explosion test. The ESR analyses were to provide further information on the effect of high energy electrons on explosives. All samples were checked for weight changes, color changes and then tested on either a heat element or an impact tester.

The heat tests were performed on a hot plate (2 types used, see Figure 8 and Figure 9), which was preheated to a temperature well above the known explosion temperature. In some tests, the glass tube containing the explosive was placed on the heat element; in other tests the explosive was first shaken onto a dish for better visual inspection before being placed on the heat element. The heat test was also employed as a back-up test for those samples which did not explode on the impact tester but had been smashed into the garnet paper disc. Specifically, the explosive was placed on the preheated element by reaching around blast-proof glass with a gloved hand holding the pliers type device which held the glass container. Explosion was always sharp, whereas

dudding was readily evident by a total lack of any sound or other evidence of rapid energy release.

Impact tests were performed on most later samples. Prior to any test of a radiated sample, an equivalent sample of non-radiated explosive was tested to insure that sample size was adequate for the set drop height. Although a mechanical impact tester (weight dropped by releasing a string which was hand-held) was used on a few explosives, most explosive samples were impact tested on the same apparatus used in Ref. [7]. This impact tester utilized an electro-magnetic release for the weight which dropped along a vertical track until it hit a steel cylinder known as the floating hammer. This hammer normally rested on the explosive under test.

The explosive was shaken onto a half-inch diameter garnet paper. This unit was then placed into a steel cup and onto a steel anvil. The latter was also removable from the cup. In order to detect the explosion without basing the decision purely on sound, a strip of paper was placed in the cup so that the interior was lined. At a setting of 100 cm, the microswitches caused release of the weight which then impacted to cause the explosive sample to be compressed and shocked. An explosion was readily visible when the paper was inspected and a dark combustion spur was seen. In case of absence of the spur, the garnet disc was removed, inspected and then the heat test was performed as a back-up test to verify the lack of explosion.

Figures 10, 11, and 12 serve to illustrate the above procedure and description. As mentioned, two electron spin resonance analyses were conducted. A qualitative analysis was performed on the last sample of lead styphnate and lead azide tested to ascertain the relationship between dudding and ESR signal. Using a VARIAN EPR Spectrometer, a minute portion of the radiated sample was used. An actual graph of the signal was obtained. The settings on the spectrometer and signal recorded on the graph produced, were then qualitatively compared to previous tests and settings, thus providing a qualitative description of "weak" signal and "no" signal. Hence, the qualitative analysis was a relative comparison based on experience.

IV. EXPERIMENTAL RESULTS

The tests will be presented in chronological order to indicate peculiarities and variations in methods. A summary table at the end of this section will present a shortened version of tests conducted and results obtained.

Test A

Twenty-five milligrams of lead styphnate were encapsulated in a glass device and exposed to a total dose of about 1×10^9 rads. The average dose rate was $1 \times 10^6 \frac{\text{rads}}{\text{sec}}$.

The radiated container with sample was not broken open. A visual inspection revealed a dark brown circular area of about 2 cm in diameter, centered on the sample. This comparatively larger cross-section indicates that enough energy was deposited in the glass to cause the color change and is attributed to the wings of the Gaussian distribution. The color change in glass was observed throughout all tests conducted and is a normal phenomenon.

The sample (confined in the glass) was heat tested on a Corning Hot Plate set at its maximum temperature of 950°F. The sample exploded and the hot plate was damaged (Figure 8).

Test B

Twenty-five milligrams of lead styphnate were loaded into a long, open end glass tube. The sample was exposed to a dose of 1×10^9 rads. The dose rate was $1.4 \times 10^6 \frac{\text{rads}}{\text{sec}}$. Figure 13 illustrates the dark circle on the glass tube and

microscope slide, which was used as a mounting in this particular test. The explosive was shaken out of the glass onto a dish for visual inspection. The sample had changed color from its normal gold to a dark rust brown color. It was noted that the sample still had a sparkling, crystalline look. This characteristic remained for all tests conducted on lead styphnate.

The dish with the radiated explosive was heat tested. The result was explosion.

Test C

Fourteen milligrams of PETN were loaded into the long glass tube. A greater amount would have resulted in a column of explosive too large for the beam cross-section. This sample was exposed to a total dose of 1×10^9 rads, as in two previous tests. The dose rate was $0.5 \times 10^6 \frac{\text{rads}}{\text{sec}}$. Weight reduction was negligible. The color of the explosive changed from white to gray. The glass tube, with explosive, was heat tested and the result was no explosion. As for previous non-radiated samples of PETN, the test sample boiled off and evaporated when placed on the red hot heat element.

Test D

Twenty-five milligrams of lead azide were loaded into the long glass tube and again exposed to 1×10^9 rads. The dose rate was $1.4 \times 10^6 \frac{\text{rads}}{\text{sec}}$. There was no weight reduction. The color of the explosive changed from its normal mustard color to a dark brown.

The glass tube, with explosive, was heat tested by direct placement onto the red hot element and exploded.

Test E

Eight milligrams of TNT were loaded into the long glass tube. Total dose was again 1×10^9 rads and dose rate was $1.5 \times 10^6 \frac{\text{rads}}{\text{sec}}$. There was no weight loss. The color of this explosive changed from its normal white to caramel.

Since non-radiated samples of TNT of this sample size could not be heat tested, the LINAC sample within its glass tube was impact tested. The result was an explosion.

Test F

The sample of HMX weighed 7.7 mg and was loaded into the long glass tube. It was exposed to 1×10^9 rads, as in the tests before, and the dose rate was $1.5 \times 10^6 \frac{\text{rads}}{\text{sec}}$.

There was no appreciable weight loss (<0.25%). The color of HMX changed from its normal white to caramel. Since non-radiated samples of HMX of this sample size could not be heat tested, the LINAC sample within its glass tube was impact tested. The result was no explosion, as ascertained by examination of the paper strip indicator.

Test G

Four milligrams of lead azide were loaded into a long glass tube. The total dose was increased to slightly over 3×10^{10} rads. The dose rate was $2.5 \times 10^6 \frac{\text{rads}}{\text{sec}}$. Again, there was no weight loss. The color changed as previously described.

The glass tube with sample was heat tested. As the color of the glass changed from dark to clear, the sample evaporated. An explosion did not result.

Test H

Test H was conducted simultaneously with Test G in that two glass tubes containing lead azide and lead styphnate were mounted back to back. Test H concentrated on 3.3 milligrams of lead styphnate loaded in a long glass tube. The sample was exposed to a total dose as for Test G; i.e. 3.2×10^{10} rads and a dose rate of $2.5 \times 10^{6} \frac{\text{rads}}{\text{sec}}$. The sample was heat tested and there was no explosion.

Test I

As for the pair of tests G and H, this test was conducted in the back to back mounting method with Test J. In this test eleven milligrams of lead azide were loaded in the long glass tube. The dose was about 2.6×10^{10} rads. The dose rate was $2.4 \times 10^{6} \frac{\text{rads}}{\text{sec}}$. There was no appreciable weight reduction. Color change was as previously described.

The explosive was shaken from the glass tube onto a dish and then heat tested. There was no explosion. Figure 9 illustrates this particular sample after having been on a red hot element in excess of 2 minutes.

Test J

The weight of the lead styphnate sample was 7.9 milligrams. It was second in line to lead azide (as in Test I). Dose and dose rate were as for Test I.

There was no weight reduction. The explosive was shaken from the long glass tube into a dish for visual inspection. Some of the lead styphnate was more difficult to get out of the glass tube and some was not as dark as the other portion from the tube. Upon heat testing, the sample exploded.

Test K

A weight of 12.8 milligrams of lead azide was loaded in the short glass tube. The total dose was about 1×10^{10} rads. The dose rate was $1 \times 10^{6} \frac{\text{rads}}{\text{sec}}$.

A weight analysis was not conducted. The color of the short glass tube was essentially black. The explosive was shaken from the glass tube into a dish and appeared to have changed its color as previously mentioned (see Figure 14).

An impact test was conducted on the radiated sample. Figure 15 indicates that there was no explosion. Further, the garnet paper disc, which contained the squashed lead azide, was heat tested and did not explode.

Test L

A weight of 10.3 milligrams of lead styphnate was loaded into a short glass tube and exposed to a total dose of 2.4×10^{10} rads. The dose rate was $2 \times 10^{6} \frac{\text{rads}}{\text{sec}}$.

A weight analysis was not conducted. The small glass tube was very black. After a cooldown to 170 milliroentgens, the sample was carried to a laboratory where a qualitative electron spin resonance revealed a weak signal. Thus only very few unpaired electrons existed; i.e. the concentration

of free electrons was not high. Since only a very minute portion of the sample was used in the ESR analysis, the remainder of the radiated explosive was shaken out onto a dish and then impact tested. Figures 16 and 17 illustrate that the result was an explosion.

Test M

A weight of 19.3 milligrams of lead azide was loaded into a short glass tube and exposed to a dose of 1.2×10^{10} rads. The dose rate was $2.5 \times 10^{6} \frac{\text{rads}}{\text{sec}}$.

A weight analysis was conducted and in this one test a reduction of about nine percent was noted. An ESR analysis was conducted. At room temperature and using the same equipment, procedure, and qualitative analysis as discussed in Test L, this sample of radiated lead azide failed to spike on the graph and thus a detectable signal, which would indicate some free electrons, was not obtained. There were no detectable unpaired electron spins.

The sample was shaken onto a dish. Some of the lead azide was darker than the remainder. In addition, difficulty was experienced in shaking the most inner explosive (lighter in color) out.

An impact test was conducted and there was no explosion. The garnet paper disc with the compressed sample was then heat tested. The result again was no explosion.

EXPLOSIVE	TEST	WEIGHT (mg.)	CONTAINER	DOSE (rads)	DOSE RATE ($\frac{rads}{sec}$)	ESR	TYPE TEST	TEST RESULT	REMARKS
Pb Azide	D	25.0	long tube	1×10^{9}	1.4×10^{6}	No	heat	explosion	
	G	4.0	long tube	3.2×10^{10}	2.5×10^{6}	No	heat	no explosion	
	I	11.0	long tube	2.6×10^{10}	2.4×10^{6}	No	heat	no explosion	
	K	12.8	short tube	1×10^{10}	1×10^{6}	No	impact	no explosion	
	M	19.3	short tube	1.2×10^{10}	2×10^{6}	Yes	impact	no explosion	ESR: No Signal
Pb Styphnate	A	25.0	capsule	1×10^{9}	1×10^{6}	No	heat	explosion	
	B	25.0	long tube	1×10^{9}	1.4×10^{6}	No	heat	explosion	
	H	3.3	long tube	3.2×10^{10}	2.5×10^{6}	No	heat	no explosion	
	J	7.9	long tube	2.6×10^{10}	2.4×10^{6}	No	heat	explosion	
	L	10.3	short tube	2.4×10^{10}	2.5×10^{6}	Yes	impact	explosion	ESR: Weak Signal
TNT	E	8.0	long tube	1×10^{9}	1.5×10^{6}	No	impact	explosion	
HMX	F	7.7	long tube	1×10^{9}	1.5×10^{6}	No	impact	explosion	
PETN	C	14.0	long tube	1×10^{9}	0.5×10^{6}	No	heat	explosion	

TABLE I.

TEST SUMMARY

V. DISCUSSION OF RESULTS

Of the thirteen tests conducted on five different explosives, the dose varied from a low of about 1×10^9 rads to a high of 3.2×10^{10} rads. The emphasis of testing was placed on the explosives lead azide and lead styphnate, whereas testing on TNT, HMX, and PETN was extremely limited. The latter three explosives, of high explosive and booster type, presented difficulty in testing methods because of both beam size limitation and laboratory tests by heat and impact. The initiator explosives lead styphnate and lead azide, in comparison, were more dense and easily testable by heat and impact. Thus, tests (five each) were concentrated on lead azide (a non-benzene ring, non-carbon chain type) and lead styphnate (benzene ring type). Both types of explosives contain lead in their molecules and were expected to react differently to radiation. This was perhaps proven by noting that eighty percent of the test results for lead azide were no explosion, whereas for lead styphnate there were explosions.

The reason why a shift was made from the long glass tube to the short glass tube was to insure a method whereby the entire holder with sample would be contained within the beam cross-section of 1 cm^2. A further effort to keep the sample within this flux area was to limit the sample size so that more assurance could be gained in getting the ϕ_{peak} onto the sample and limiting the non-desirable extension of the sample

column into the wings of the Gaussian distribution. However, because of sample holder and type of explosive, weights of 4.0 mg and 3.3 mg (Tests G and H respectively) are essentially at the minimum which is testable. This philosophy applied to the TNT, HMX, and PETN sample weights, (as in Tests E, F, and C) and was degrading to the test results; i.e. larger samples should have been used for these explosives to permit impact testing. Therefore, the test results for TNT, HMX, and PETN, should not be considered indicative as to their reaction to high energy radiation, using heat or impact testing.

The test results of greatest interest are those obtained from lead azide and lead styphnate. Table I indicates that lead azide was completely desensitized for four out of five tests. The first test on lead azide (Test D) resulted in an explosion. This result could have been caused by the fact that some non-radiated lead azide had clung to the walls of the long glass and it was sufficient to cause the explosion. Also, this sample received a somewhat smaller dose than the later samples.

Four out of five tests with lead styphnate resulted in explosions. The one no explosion result could be attributed to the extremely small sample size (3.3 mg), the highest dose (3.2×10^{10} rads), or a combination of the two reasons.

Tests G and H were conducted in the back to back method, as were Tests I and J. The effect of high energy electron penetration and ionization is not significantly changed by

mounting a glass tube, with the sample, directly behind another, as long as the beam centers on both explosive samples. Practically, the back to back method is more expedient and cuts the time of LINAC usage; however, the penalty is lower reliability in alignment of samples. The sample sizes for Tests G and H were 4.0 and 3.3 milligrams. It was interesting to note that the 3.3 mg of lead styphnate was contrary in test behavior to the other samples of lead styphnate. In general, these sizes should be considered too small and it is felt that the test reliability is less than for the others.

Tests I and J were conducted to verify test results of the G and H tests. The weights of lead azide and lead styphnote were more than doubled but the total dose was a little less.

The shift to the short glass tube container and impact testing (which permitted the additional heat test as a second test in case of no explosion by impact) provided the best method of testing. Thus, Tests K, L, and M were those providing the most reliable data. For lead azide, Tests K and M added validity to two previous no explosion results. In the case of lead styphnate, Test L added validity to three out of four explosion results.

The electron spin resonance (ESR) tests were not considered until late during this study. The hope was that a relationship between unpaired electrons (signal) and explosion/no explosion could be established. The ESR test results

of no signal for lead azide and a weak signal for lead styphnate do not indicate such a connection.

In summary, doses of the order of 10^{10} rads dudded lead azide and had no such effect on lead styphnate. The test results on TNT, HMX, and PETN should be regarded as limited in value since the difficulty of testing dudding, in the small sample sizes which were radiated, made a good criterion virtually impossible.

VI. CONCLUSIONS AND RECOMMENDATIONS

A. The bond energy theory was supported by the behavior of the lead azide and lead styphnate molecules when subjected to a high energy electron beam. Specifically, the peculiarity of resistance to radiation as explained by the Sponge Effect seemed demonstrated by the test results of lead styphnate. Having a benzene ring and an ionic bond configuration, energy of up to 3.20×10^{10} rads was ineffective in the macroscopic behavior of lead styphnate. On the contrary, the radiation did affect the non-benzene ring configuration of lead azide. It is recommended that an ESR be conducted on radiated samples of lead azide to find out if a signal is detectable from lowest to highest dosage. Furthermore, it is recommended that a chemical analysis be conducted to ascertain products, which may support a suspicion that the lead azide molecule was virtually changed to pure nitrogen and lead when subjected to radiation.

B. Ionizing radiation from 1×10^{9} rads to 3.2×10^{10} rads will cause lead azide to be completely desensitized and have no effect on the macroscopic behavior of lead styphnate when subjected to either heat or impact tests.

C. Data on the effects of high energy electrons on TNT, HMX and PETN was insufficient to form a conclusion.

D. It is recommended that if future tests of this type are conducted, sample sizes should be twenty milligrams for best impact testing and that impact tests be utilized to verify the impact sensitivity of standard explosives when subjected to various doses. Furthermore, the theory and correlation of dose rate should be considered.

APPENDIX A

EXPLOSIVE YIELD CALCULATION

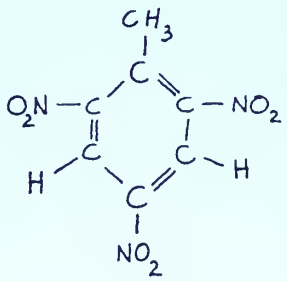
The energy released by an explosion is not only due to bond energy but also from a process of residual solids to gaseous products when unlimited oxygen is present.

Reference [11] and Ref. [6] are specific on several methods of explosive power calculations and the associated theory and assumptions. The method taught at the United States Naval Postgraduate School is a means whereby several characteristics can be relatively easily calculated.

The explosive will proceed from a solid to a gas resulting in an energy based on complete combustion in unlimited oxygen (ΔE_C). This energy is equal to the sum of energy released by use of internal oxygen only (ΔE_E) and the energy released when residual solids such as carbon combines with oxygen to form carbon dioxide ($\Delta E_{products}$). In the entire process, the prioity of oxygen use is:

1. oxygen $\xrightarrow{H}$ H_2O
2. oxygen $\xrightarrow{C}$ CO_2

A sample yield calculation for the explosive TNT is as follows:



TNT

Energy bonds:

Type	No.	ΔE_C each	Total ΔE_C per bond type
C-H	5	-52.44	-262.00
C-C	4	-49.79	-199.00
C=C	3	-116.43	-349.50
$C-NO_2$	3	-16.66	-50.00
Resonance	1	+40.00	+40.00
		Total ΔE_C =	-820.50 $\frac{\text{K-cal.}}{\text{Mole}}$

With the assumption that oxygen is used as previously stated,

Explosive Reaction:

$$C_7H_5O_6N_3 \longrightarrow \frac{5}{2} H_2O_{(g)} + \frac{7}{4} CO_{2(g)} + \frac{3}{2} N_{2(g)} + \frac{21}{4} C_{(s)}$$

The change in volume $\Delta V \equiv$ sum of all the gaseous coefficients; thus,

$$\Delta V = 5.75$$

Since carbon is a residue $(\frac{21}{4})$, then it will combine with oxygen to form $CO_{2(g)}$. Referring to Ref. [11], a calculation for the change in energy due to products is:

$$\Delta E_{products} = \frac{21}{4} \times (-94.4) = -495.50 \frac{\text{K-cal.}}{\text{Mole}}$$

Since $\Delta E_C = \Delta E_E + \Delta E_{products}$

$$\Delta E_E = \Delta E_C - \Delta E_{products}$$

$$\Delta E_E = -820.50 - (-495.5) \frac{\text{K-cal.}}{\text{Mole}}$$

$$\Delta E_E = -325.0 \frac{\text{K-cal.}}{\text{Mole}}$$

Since ΔE_E was previously defined as being based on internal oxygen use, ΔE_E represents the chemical bond energy liberated:

$$Q = -325. \frac{\text{K-cal}}{\text{Mole}} \times \frac{1 \text{ mole}}{227 \text{ grams}} = 1432 \frac{\text{cal}}{\text{g}} \text{ TNT}$$

or, since 1 cal = 4.186 joule

1 joule = 10^7 ergs

then,

$$Q = 1432 \frac{cal}{g} \times \frac{4.186 \text{ joule}}{1 \text{ cal}} \times \frac{10^7 \text{ ergs}}{1 \text{ joule}} = 6.0 \times 10^{10} \frac{\text{ergs}}{\text{gm}} \text{ TNT}$$

On this bond energy and certain constants, the detonation pressure and velocity are fully dependent:

Defining the number of moles of detonation products as the ratio of the change of volume to the molecular weight of TNT,

$$N \equiv \frac{\Delta V}{M.W} = \frac{5.75}{227} = 0.0253 \frac{\text{moles}}{\text{gm}}$$

Defining the average molecular weight of the gaseous detonation products as the summation of the products as follows:

$$M \equiv \sum \left[\frac{\Delta E_E \text{ gas coefficients}}{\Delta V} \right] \times [\text{M.W. of gas}]$$

$$M = \left(\frac{5/2}{5.75}\right) \times (\text{M.W. of } H_2O) + \left(\frac{7/4}{5.75}\right) \times (\text{M.W of } CO_2) + \left(\frac{3/2}{5.75}\right) \times (\text{M.W of } N_2)$$

$$M = \frac{5/2}{5.75} \times 18 + \frac{7/4}{5.75} \times 44 + \frac{3/2}{5.75} \times 28$$

$$M = 28.5 \text{ gm}$$

Defining a constant as follows,

$$\phi = N M^{1/2} Q^{1/2}$$

$$\phi = (25.3 \times 10^{-3}) \times \sqrt{28.5} \times 1432$$

$$\phi = 5.11$$

Detonation pressure:

$$P = K \rho_o^2 \phi$$

where K $\equiv$ constant = 15.58

ρ_o = density of TNT

$P = 15.58 \times (1.65)^2 \times 5.11$

$P = \underline{217 \text{ kilobars}}$

Detonation velocity:

$D = A\,\phi^{1/2} \times (1 + B\rho_o)$

where A $\equiv$ constant = 1.01

B $\equiv$ constant = 1.30

$D = 1.01 \times \sqrt{5.11} \times [1 + 1.30 \times 1.65]$

$D = 7.18 \ \underline{\text{mm.}}$
$\underline{\text{microsec.}}$

APPENDIX B

ILLUSTRATIONS

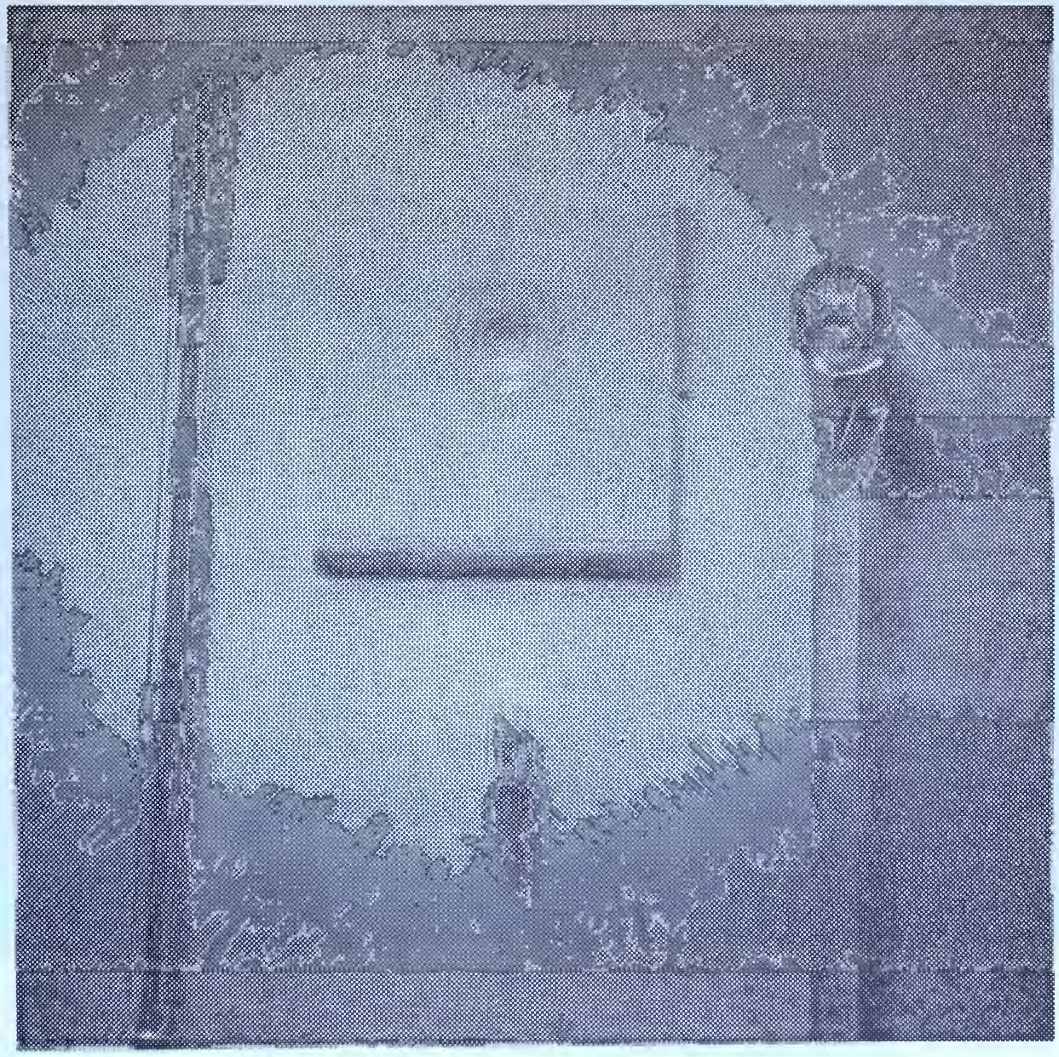


Figure 3. Glass explosive containers and dipper.

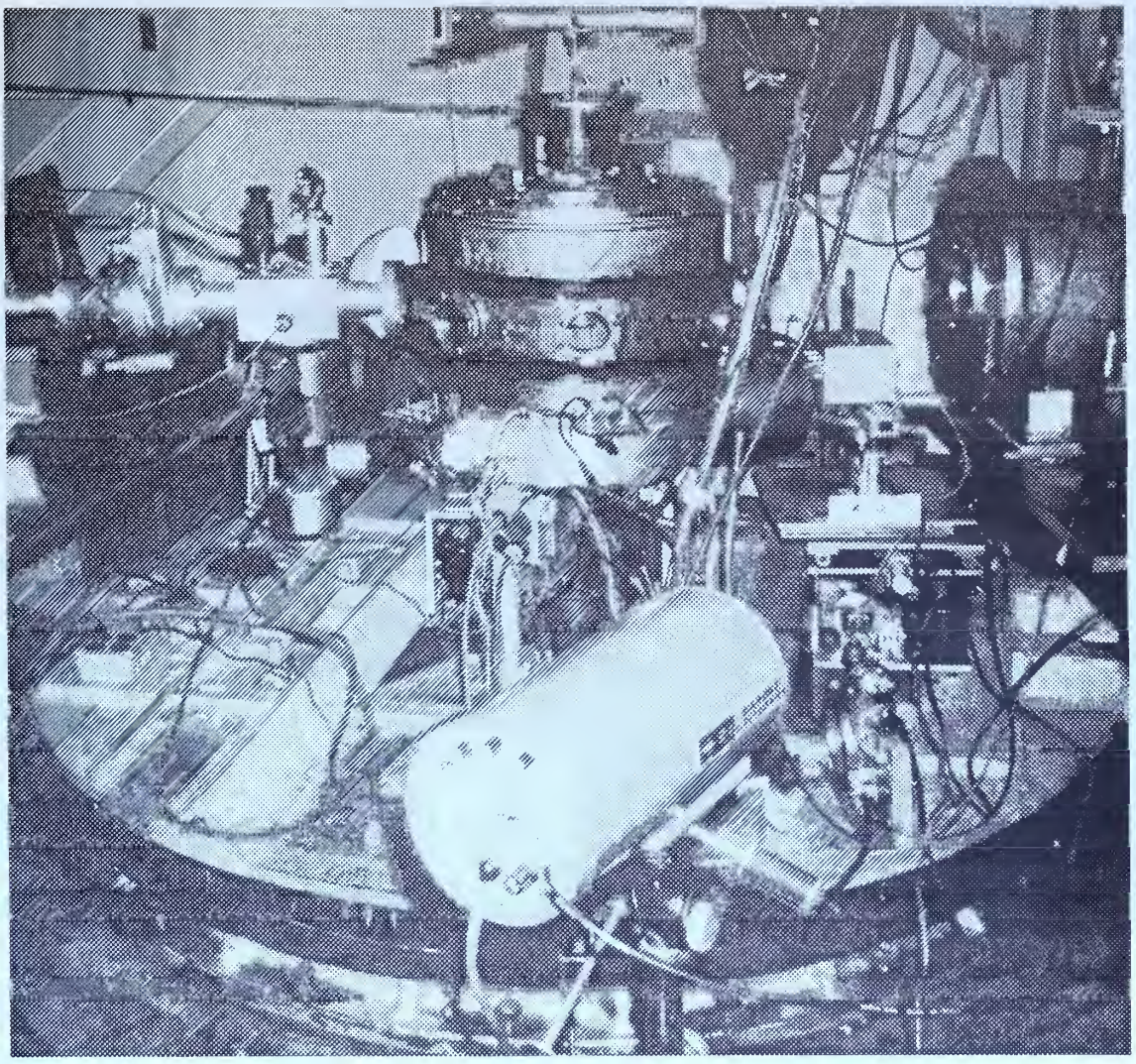


Figure 4. Accelerator end station arrangement.

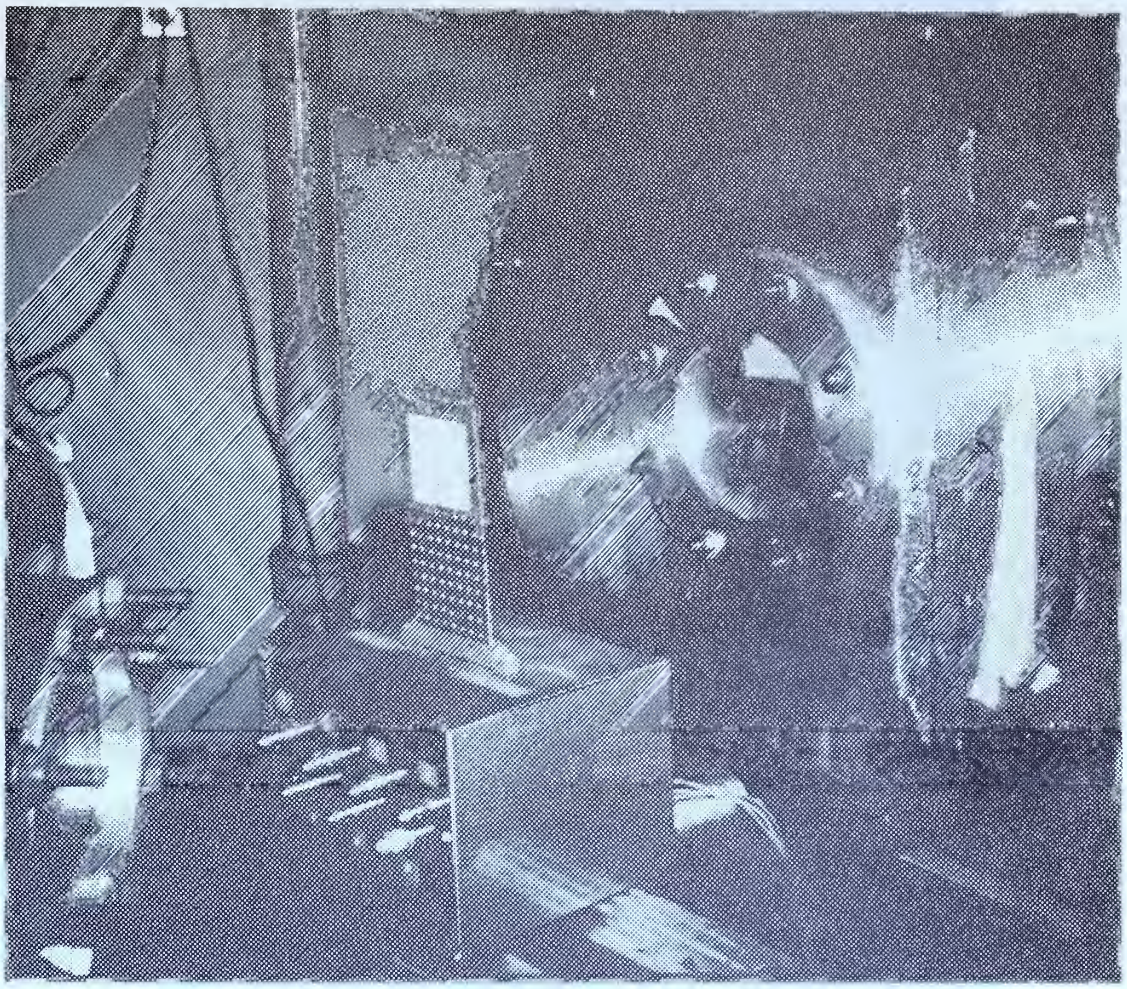


Figure 5. Target card mounting and Faraday cup.

Figure 6. T.V. view of beam on sample (short glass tube).

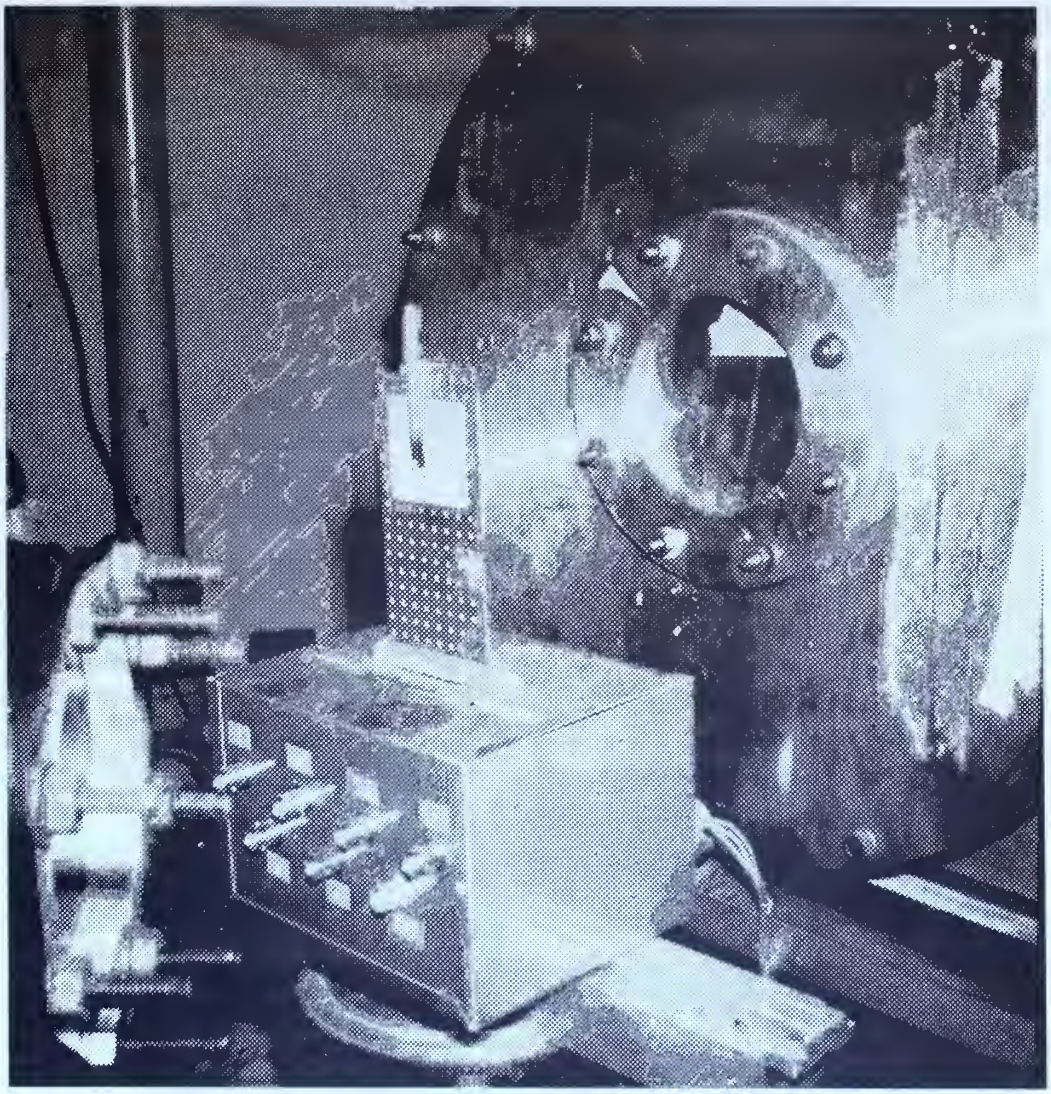


Figure 7. Mounted sample (in long glass) after radiation.

Figure 8. Corning Hot Plate (holed by sample).

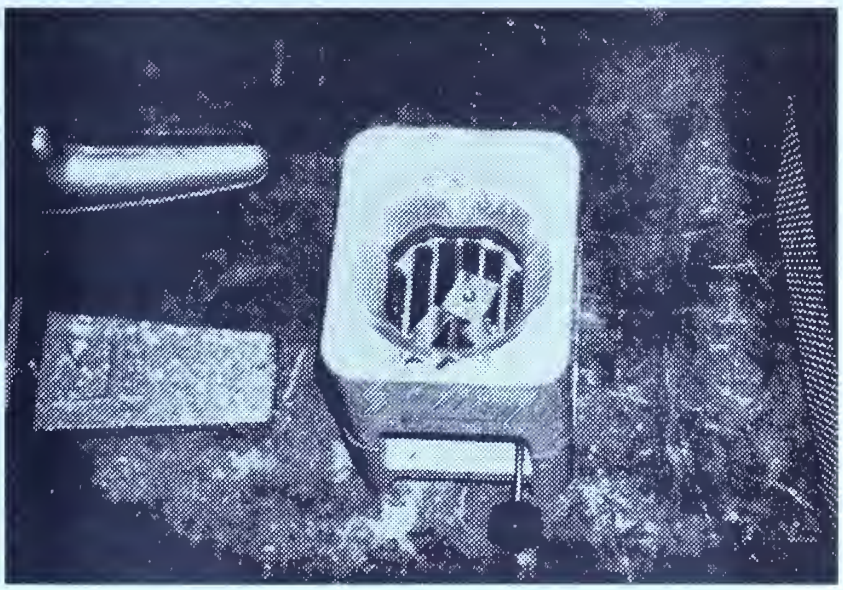


Figure 9. Boekel Hot Plate (exposed coils).

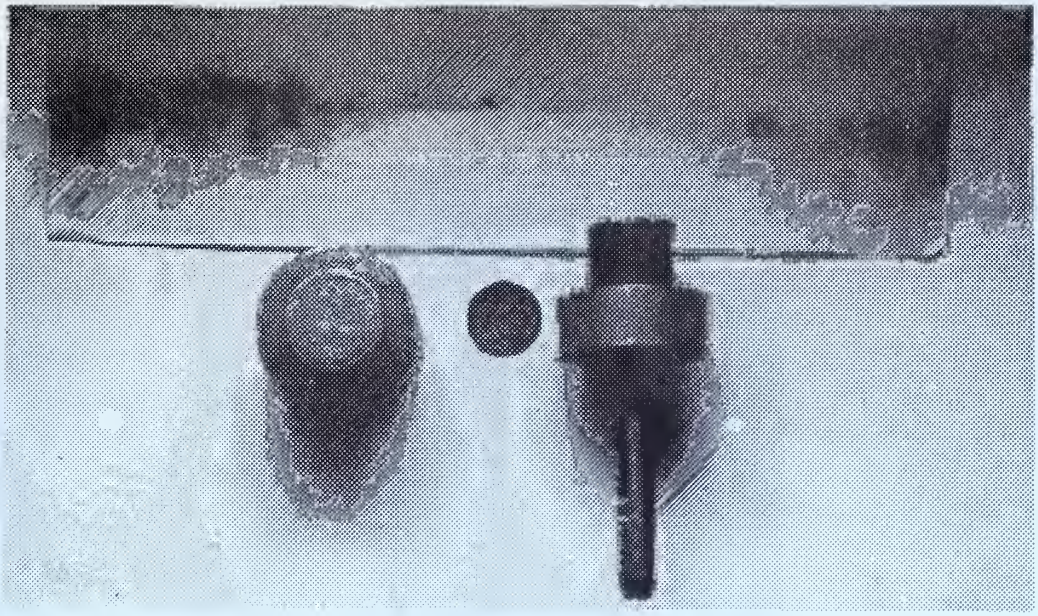


Figure 10. Impact tester parts; indicator (showing spur), floating hammer, garnet paper disc, and anvil.

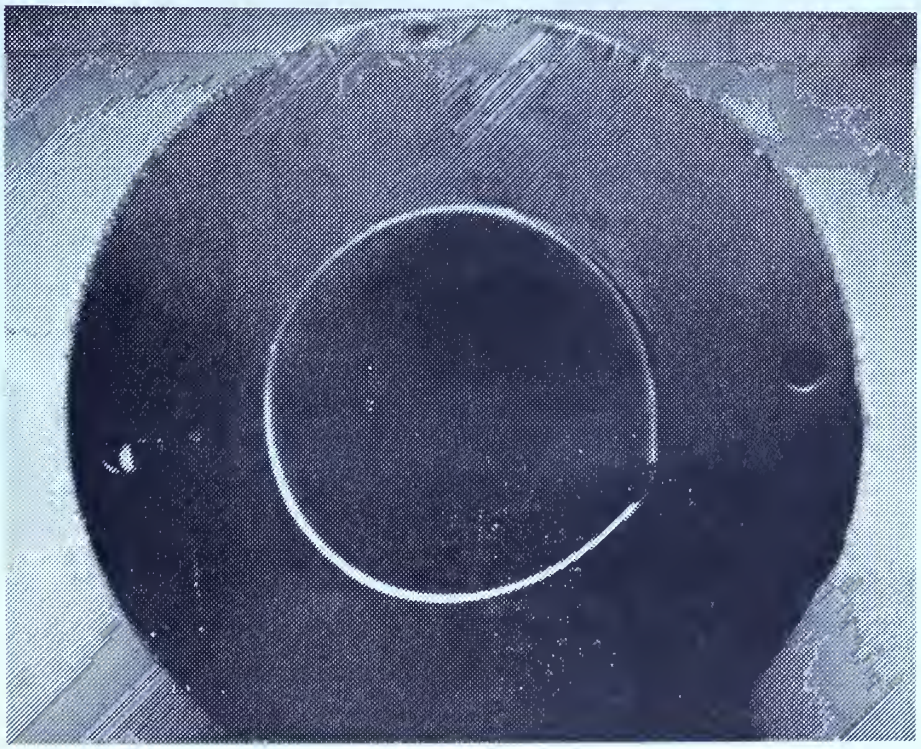


Figure 11. Impact tester steel cup with indicator in place.

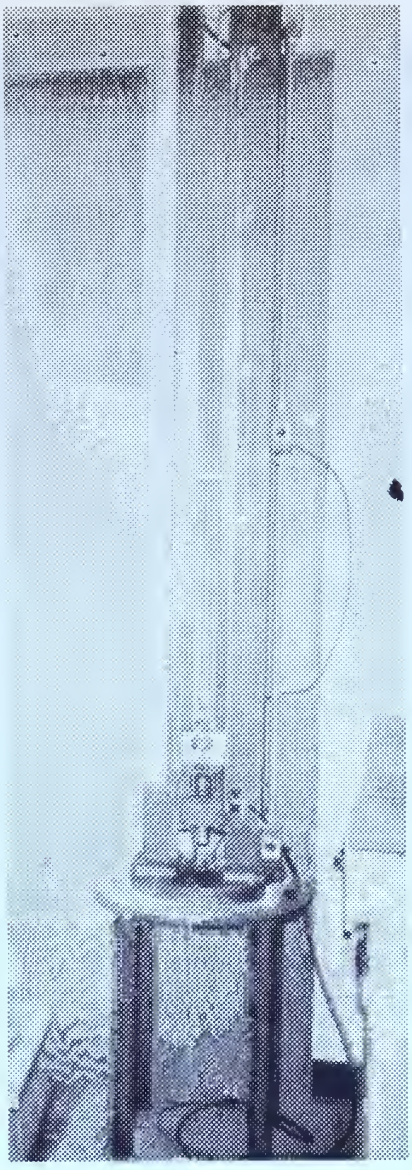


Figure 12. Impact Tester.

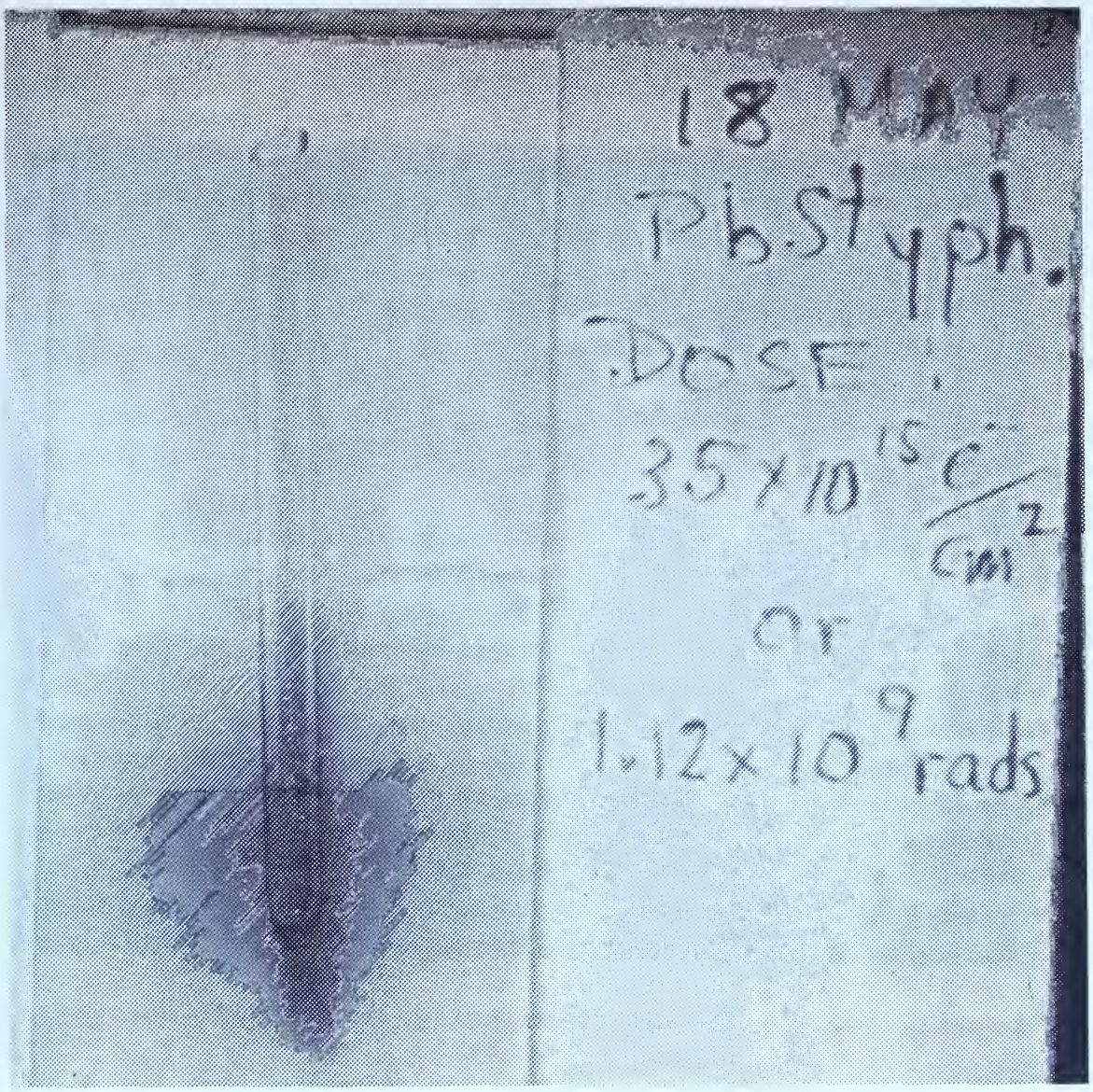


Figure 13. Long glass tube, mounted on microscope slide, after radiation.

Figure 14. Lead azide after radiation in a short glass tube and shaken onto a glass dish.

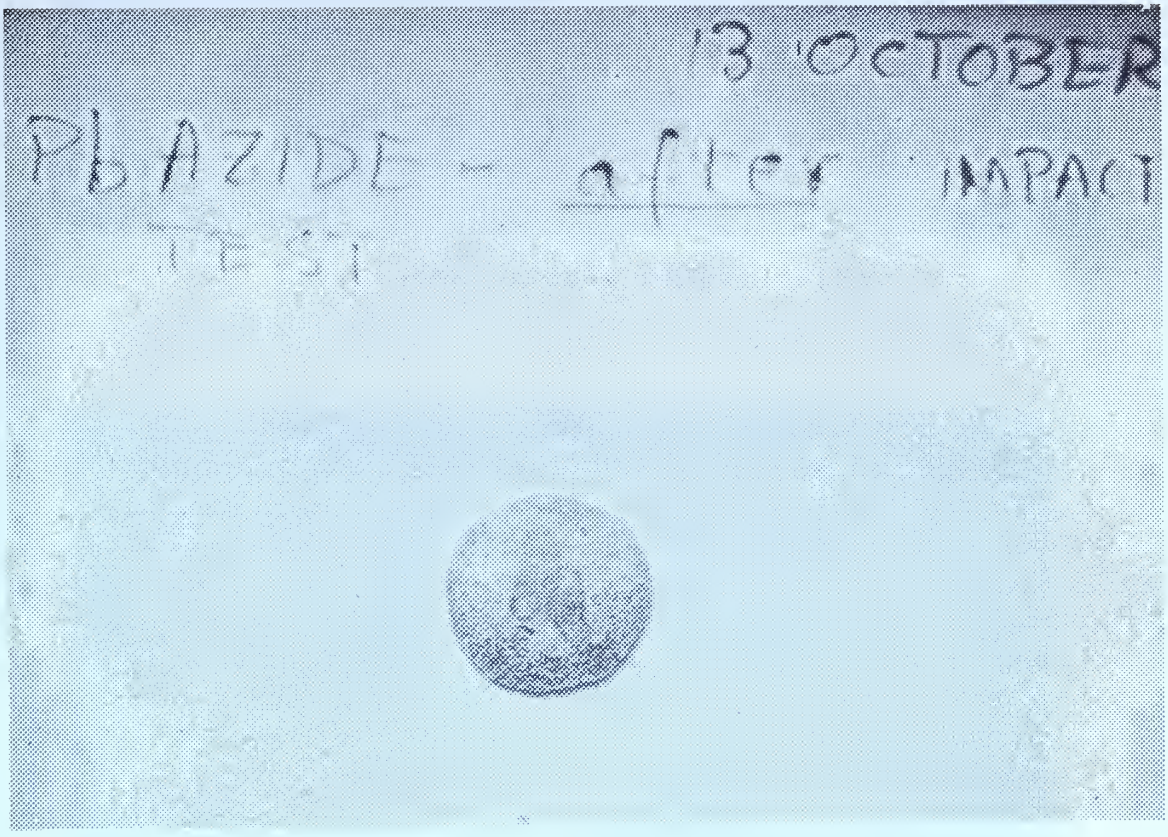


Figure 15. Lead azide on garnet paper disc after impact test.

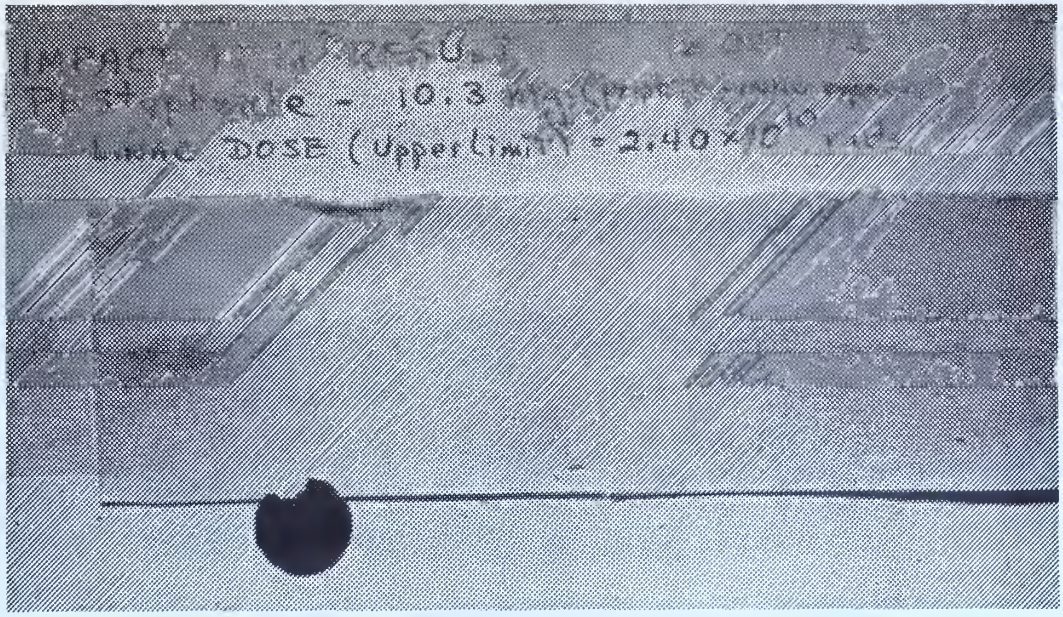


Figure 16. Impact test result of explosion.

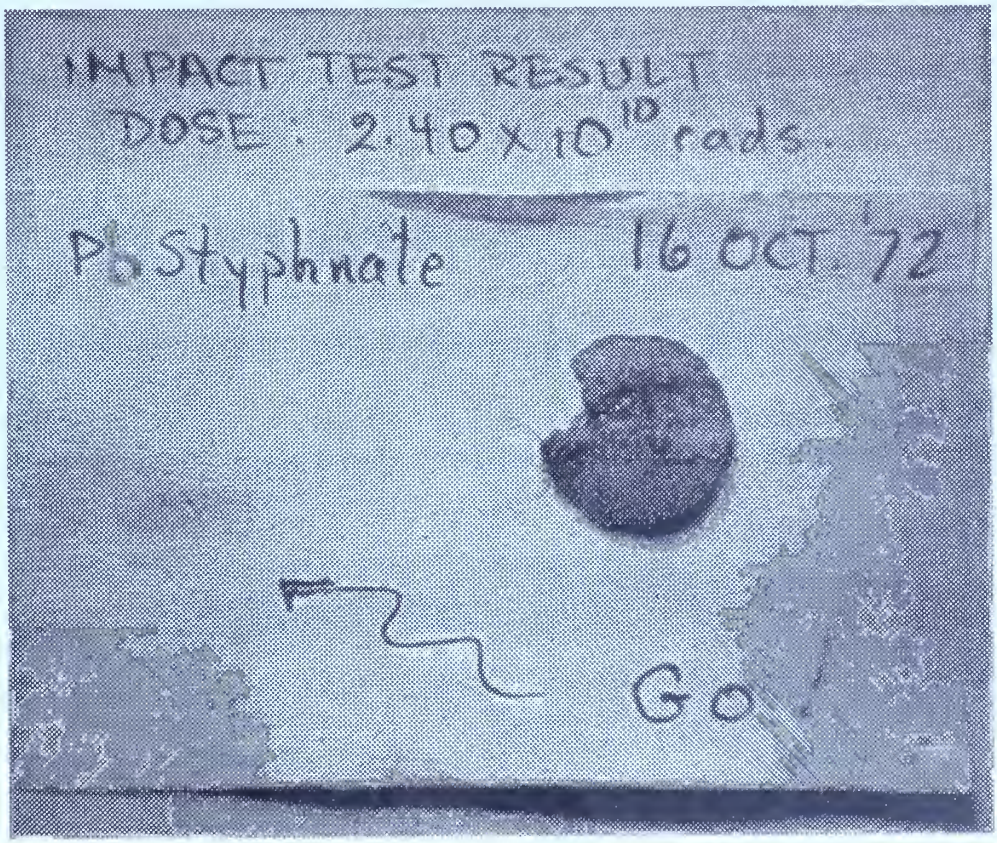


Figure 17. Close-up view of indicator and garnet disc.

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13. ABSTRACT

The effect of a sixty million electron volt beam, produced by a linear accelerator and focused on standard explosives, varying in weight from three to twenty-five milligrams, has been studied. Doses in the range from 1×10^9 rads to 3.2×10^{10} rads completely desensitized lead azide but had no effect on the explosive behavior of lead styphnate. The desensitization tests on explosives consisted of both heat and impact test methods.

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Radiation Effects on Explosives						
Electron Radiation of Explosives						
Desensitization of Explosives						
Linear Accelerator and Explosives						

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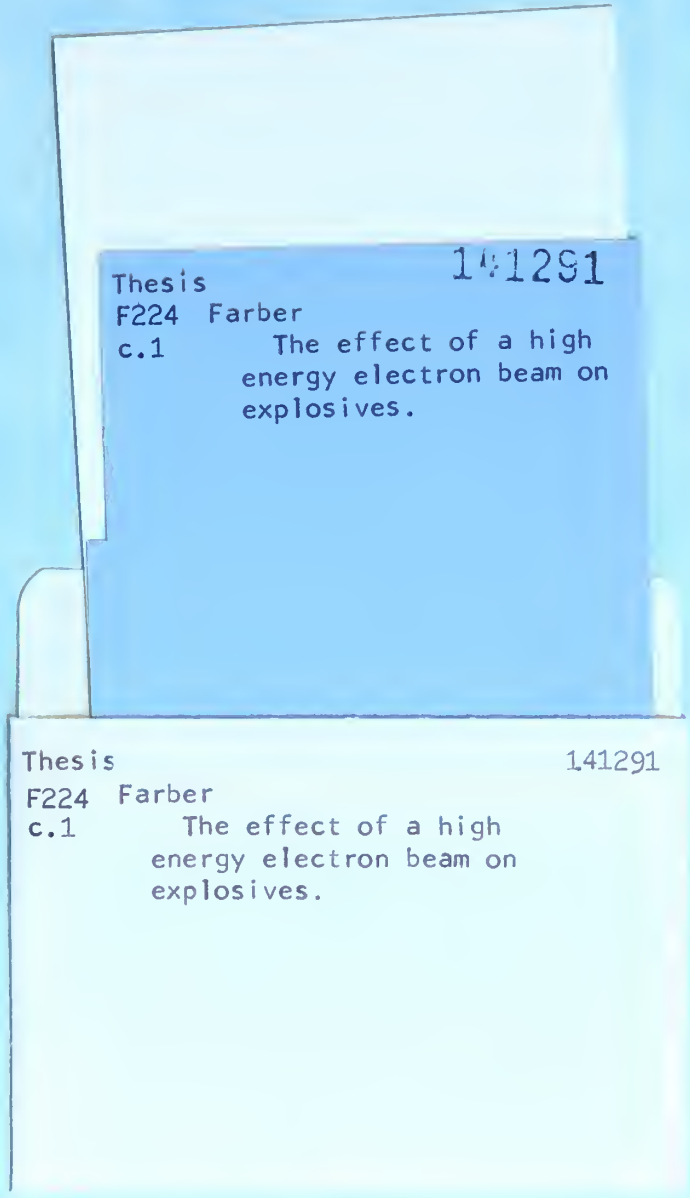

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